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14. ABSTRACT This objective of this project is to seek novel quantum phenomena in perovskite ruthenates, study their underlying physics, and explore their possible application. We have studied metal-insulator transitions in doped Ca <sub>2</sub> RuO <sub>4</sub> and Ca <sub>3</sub> Ru <sub>2</sub> O <sub>7</sub> . We find that the metal-insulator (MI) transitions in both materials can be effectively controlled by Ti and Fe doping. In Ca <sub>2</sub> RuO <sub>4</sub> we have tuned the MI transition to a transition close to a 2nd-order transition with T <sub>c</sub> ~ 250-320K and TCR <sub>max</sub> ~ 0.5 K <sup>-1</sup> (TCR: temperature coefficient of resistance). Such a MI transition is likely					
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## Report Title

Searching for exotic bulk and interfacial quantum phenomena of perovskite ruthenates

### ABSTRACT

This objective of this project is to seek novel quantum phenomena in perovskite ruthenates, study their underlying physics, and explore their possible application. We have studied metal-insulator transitions in doped  $\text{Ca}_2\text{RuO}_4$  and  $\text{Ca}_3\text{Ru}_2\text{O}_7$ . We find that the metal-insulator (MI) transitions in both materials can be effectively controlled by Ti and Fe doping. In  $\text{Ca}_2\text{RuO}_4$  we have tuned the MI transition to a transition close to a 2nd-order transition with  $T_c \sim 250\text{-}320\text{K}$  and  $\text{TCR}_{\text{max}} \sim 0.5\text{ K}^{-1}$  (TCR: temperature coefficient of resistance). Such a MI transition is likely useful for developing a new generation of bolometric detection technology. In addition, we have investigated electronic and magnetic properties of  $(\text{Sr}_{1-x}\text{Ca}_x)_3\text{Ru}_2\text{O}_7$ . We find that this system exhibits complex electronic and magnetic ground states and that magnetic states are strongly coupled with electronic states. This demonstrates the strong interplay between the charge and spin degrees of freedom in ruthenates. This finding advances our understanding of strongly correlated phenomena in oxides.

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### List of papers submitted or published that acknowledge ARO support during this reporting period. List the papers, including journal references, in the following categories:

#### (a) Papers published in peer-reviewed journals (N/A for none)

“Unusual heavy-mass nearly ferromagnetic state with a surprisingly large Wilson ratio in the double layered ruthenates  $(\text{Sr}_{1-x}\text{Ca}_x)_3\text{Ru}_2\text{O}_7$ ”,

Z. Qu, L. Spinu, H.Q. Yuan, V. Dobrosavljevic, W. Bao, J.W. Lynn, M. Nicklas, J. Peng, T.J. Liu, D. Fobes, E. Flesch, and Z.Q. Mao, Phys. Rev. B 78, 180407 (Rap. Com.) (2008).

“Complex electronic states in double layered ruthenates  $(\text{Sr}_{1-x}\text{Ca}_x)_3\text{Ru}_2\text{O}_7$ ”,

Z. Qu, J. Peng, T.J. Liu, D. Fobes, L. Spinu, and Z.Q. Mao, Phys. Rev. B 80, 115130 (2009).

Number of Papers published in peer-reviewed journals: 2.00

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#### (b) Papers published in non-peer-reviewed journals or in conference proceedings (N/A for none)

Number of Papers published in non peer-reviewed journals: 0.00

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#### (c) Presentations

Probing orbital-dependent magnetism in layered perovskite ruthenates through angle-dependent magnetoresistivity

D. Fobes, T. J. Liu, Z. Qu, H.Q. Yuan, M. Salamon, M. Zhou, J. Hooper, Z. Q. Mao.

American Physical Society March meeting 2009, Pittsburgh.

Antiferromagnetism and bulk spin valve effect in  $\text{Ca}_3(\text{Ru}_{1-x}\text{Ti}_x)_2\text{O}_7$

J. Peng, T.J. Liu, Z. Qu, E. Vehstedt, B. Qian, D. Fobes, L. Spinu, W. Bao, and Z.Q. Mao

American Physical Society March meeting 2009, Pittsburgh.

Electronic phase diagram in double layered ruthenates  $(\text{Sr}_{1-x}\text{Ca}_x)_3\text{Ru}_2\text{O}_7$

Z. Qu, J. Peng, T.J. Liu, D. Fobes, B. Qian, L. Spinu, and Z.Q. Mao

American Physical Society March meeting 2009, Pittsburgh.

Number of Presentations: 3.00

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#### Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Number of Non Peer-Reviewed Conference Proceeding publications (other than abstracts): 0

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#### Peer-Reviewed Conference Proceeding publications (other than abstracts):

(d) Manuscripts

Strong interplay between the lattice and spin degrees of freedom  
in (Sr1?xCax)3Ru2O7

Jin Peng,Zhe Qu,Bin Qian,David Fobes,Tijiang Liu,  
Xiaoshan Wu, H. M. Pham, Leonard Spinu, and Z.Q. Mao,  
Submitted to Physical Review B

Number of Manuscripts:1.00

Number of Inventions:

Graduate Students

<u>NAME</u>	<u>PERCENT SUPPORTED</u>
David Fobes	0.30
Bin Qian	0.20
FTE Equivalent:	0.50
Total Number:	2

Names of Post Doctorates

<u>NAME</u>	<u>PERCENT SUPPORTED</u>
FTE Equivalent:	
Total Number:	

Names of Faculty Supported

<u>NAME</u>	<u>PERCENT SUPPORTED</u>	National Academy Member
Zhiqiang Mao	0.40	No
FTE Equivalent:	0.40	
Total Number:	1	

Names of Under Graduate students supported

<u>NAME</u>	<u>PERCENT SUPPORTED</u>
Erin	
Erin Vehsted	0.10
FTE Equivalent:	0.10
Total Number:	2

### Student Metrics

This section only applies to graduating undergraduates supported by this agreement in this reporting period

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### Names of Personnel receiving masters degrees

NAME

Total Number:

### Names of personnel receiving PHDs

NAME

Total Number:

### Names of other research staff

NAME

PERCENT SUPPORTED

FTE Equivalent:

Total Number:

### Sub Contractors (DD882)

### Inventions (DD882)



## Final Technical Report

Project title: Searching for exotic bulk and interfacial quantum phenomena of perovskite ruthenates

Award Number: W911NF-08-C-0131

Principal investigator: Zhiqiang Mao, Tulane University

### Abstract:

This objective of this project is to seek novel quantum phenomena in perovskite ruthenates, study their underlying physics, and explore their possible application. We have studied metal-insulator transitions in doped  $\text{Ca}_2\text{RuO}_4$  and  $\text{Ca}_3\text{Ru}_2\text{O}_7$ . We find that the metal-insulator (MI) transitions in both materials can be effectively controlled by Ti and Fe doping. In  $\text{Ca}_2\text{RuO}_4$  we have tuned the MI transition to a transition close to a 2<sup>nd</sup>-order transition with  $T_c \sim 250\text{-}320\text{K}$  and  $\text{TCR}_{\text{max}} \sim 0.5 \text{ K}^{-1}$  (TCR: temperature coefficient of resistance). Such a MI transition is likely useful for developing a new generation of bolometric detection technology. In addition, we have investigated electronic and magnetic properties of  $(\text{Sr}_{1-x}\text{Ca}_x)_3\text{Ru}_2\text{O}_7$ . We find that this system exhibits complex electronic and magnetic ground states and that magnetic states are strongly coupled with electronic states. This demonstrates the strong interplay between the charge and spin degrees of freedom in ruthenates. This finding advances our understanding of strongly correlated phenomena in oxides.

### Summary of achievements:

Strongly correlated oxides have been the subject of intense study for the last two decades. Many of these materials exhibit exciting and technically useful properties; examples include high temperature superconductivity, colossal magnetoresistance, ferromagnetism and ferroelectricity. Recently, perovskite ruthenates  $(\text{Sr,Ca})_{n+1}\text{Ru}_n\text{O}_{3n+1}$  have become a focus in this field, since they exhibit a rich variety of fascinating ordered ground states, such as spin-triplet superconductivity, itinerant magnetism, orbital ordering, Mott insulator, and a field-tuned electronic nematic phase. The close proximity of these exotic states testifies to the delicate balance among the charge, spin, lattice and orbital degrees of freedom in ruthenates, and provides a remarkable opportunity for observing novel quantum phenomena through controlling external stimuli and potential applications. This objective of this project is to seek novel quantum phenomena in ruthenates, study their underlying physics, and explore their possible application. We have made significant progresses as summarized in the following:

#### 1) Metal-insulator transitions near room temperature in Sr-,Ti-, and Fe-doped $\text{Ca}_2\text{RuO}_4$

Uncooled bolometric detectors are a key technology for the military, useful for night vision for soldiers. A microbolometer requires a temperature sensitive element or thermometer that displays a large rate of change of resistance with respect to temperature,  $dR/dT$  [1]. The responsivity and detectivity of a bolometer are sensitively dependent on the relative magnitude of the change in resistance, *i.e.*,  $(1/R)dR/dT$ , known as the temperature coefficient of resistance (TCR), and on the  $1/f$  noise voltage. Current practical microbolometers are mainly based on amorphous silicon (*a*-Si) and vanadium oxides ( $\text{VO}_x$ ) [1]. Their TCR values are in the range of  $0.01\text{-}0.05 \text{ K}^{-1}$  [1,2], which is low and limits the sensitivity of the detector. Our results obtained from this project show that doped calcium ruthenate  $\text{Ca}_2\text{RuO}_4$  is a promising candidate for bolometric detection. We have shown the MI transition in this material is tunable through chemical doping. The transition temperature can be tuned to room temperature and large TCR values ( $\sim 0.5 \text{ K}^{-1}$ ) can be obtained near the MI transition. For a MI transition to be useful in bolometric detection, it needs to be a second order transition. Our results show that this can be achieved through Fe doping. In addition, we find that the resistivity within the transition regime is tunable to a range which allows for larger bias current for these materials, which is also essentially important to raise the detectivity of a bolometer. More importantly, ruthenates allow for homogeneous epitaxial thin film growth [3,4]; the  $1/f$  noise of ruthenate films is expected to be small. These results build the base for the PI's current DEPSCoR project.

#### 2) Complex electronic and magnetic ground states of $(\text{Sr}_{1-x}\text{Ca}_x)_3\text{Ru}_2\text{O}_7$

We have studied electronic and magnetic properties of  $(\text{Sr}_{1-x}\text{Ca}_x)_3\text{Ru}_2\text{O}_7$ . We find that the magnetic ground state of  $(\text{Sr}_{1-x}\text{Ca}_x)_3\text{Ru}_2\text{O}_7$  is complex, ranging from an itinerant metamagnetic state, to an unusual heavy-mass nearly ferromagnetic (FM) state, and finally to an antiferromagnetic (AFM) state [5]. We have also elucidated the electronic properties for these magnetic states, and show that the electronic and magnetic properties are strongly coupled in this system [6]. The electronic ground state evolves from an AFM quasi-two-dimensional metal for  $x=1.0$  to an Anderson localized state for  $0.4 \leq x \leq 1.0$  (the AFM region). When the magnetic state undergoes a transition from the AFM to the nearly FM state, the electronic ground state switches to a weakly localized state induced by magnetic scattering for  $0.25 \leq x < 0.4$ , and then to a magnetic metallic state  $0.08 < x < 0.25$ . The system eventually transforms into a Fermi-liquid ground state when the magnetic ground state enters the itinerant metamagnetic state for  $x < 0.08$ . When  $x$  approaches the critical composition ( $x \sim 0.08$ ), the Fermi-liquid temperature is suppressed to zero Kelvin, and non-Fermi-liquid behavior is observed. These results demonstrate the strong interplay between the charge and spin degrees of freedom in ruthenates, which advances our understanding of strongly correlated phenomena in oxides.

#### Publications:

We have published two papers for this project.

*“Unusual heavy-mass nearly ferromagnetic state with a surprisingly large Wilson ratio in the double layered ruthenates  $(\text{Sr}_{1-x}\text{Ca}_x)_3\text{Ru}_2\text{O}_7$ ”*,

Z. Qu, L. Spinu, H.Q. Yuan, V. Dobrosavljevic, W. Bao, J.W. Lynn, M. Nicklas, J. Peng, T.J. Liu, D. Fobes, E. Flesch, and Z.Q. Mao, Phys. Rev. B **78**, 180407 (Rap. Com.) (2008).

*“Complex electronic states in double layered ruthenates  $(\text{Sr}_{1-x}\text{Ca}_x)_3\text{Ru}_2\text{O}_7$ ”*,

Z. Qu, J. Peng, T.J. Liu, D. Fobes, L. Spinu, and Z.Q. Mao, Phys. Rev. B **80**, 115130 (2009).

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[6] Z. Qu, J. Peng, T.J. Liu, D. Fobes, L. Spinu, and Z.Q. Mao, “Complex electronic states in double layered ruthenates  $(\text{Sr}_{1-x}\text{Ca}_x)_3\text{Ru}_2\text{O}_7$ ”, Phys. Rev. B **80**, 115130 (2009).